

COPY

Form PTO-1300 (Rev. 12-28-98)		US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NO. H 3722 PCT/US	
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (If known, use 37 CFR 1.5) 097807165	
INTERNATIONAL APPLICATION NO. PCT/EP99/07274		INTERNATIONAL FILING DATE October 1, 1999		PRIORITY DATE CLAIMED October 10, 1998	
TITLE OF INVENTION ADDITIVES FOR PROCESSING PLASTICS					
APPLICANT(S) FOR DO/EO/US Peter Daute, Wolfgang Seiter, Joerg-Dieter Klamann, Peter Wedl					
Applicant herewith submits to the United States Designated/Elected Office (EO/DO/US) the following items and other information:					
1.	<input checked="" type="checkbox"/>	This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.			
2.	<input type="checkbox"/>	This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.			
3.	<input type="checkbox"/>	This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39 (1).			
4.	<input checked="" type="checkbox"/>	A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.			
5.	<input checked="" type="checkbox"/>	A copy of the International Application as filed (35 U.S.C. 371(c)(2)).			
6.	<input type="checkbox"/>	is transmitted herewith (required only if not transmitted by the International Bureau).			
7.	<input checked="" type="checkbox"/>	has been transmitted by the International Bureau.			
8.	<input type="checkbox"/>	is not required, as the application was filed in the United States Receiving Office (RO/US).			
9.	<input checked="" type="checkbox"/>	A translation of the International Application into English (35 U.S.C. 371(c)(2)).			
10.	<input checked="" type="checkbox"/>	Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))			
11.	<input type="checkbox"/>	are transmitted herewith (required only if not transmitted by the International Bureau).			
12.	<input type="checkbox"/>	have been transmitted by the International Bureau.			
13.	<input type="checkbox"/>	have not been made; however, the time limit for making such amendments has NOT expired.			
14.	<input checked="" type="checkbox"/>	have not been made and will not be made.			
15.	<input type="checkbox"/>	A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).			
16.	<input checked="" type="checkbox"/>	An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). (UNEXECUTED)			
17.	<input type="checkbox"/>	A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).			
Items 11. to 16. below concern other document(s) or information included:					
18.	<input type="checkbox"/>	An Information Disclosure Statement under 37 CFR 1.97 and 1.98.			
19.	<input type="checkbox"/>	An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.			
20.	<input checked="" type="checkbox"/>	A FIRST preliminary amendment			
21.	<input type="checkbox"/>	A SECOND or SUBSEQUENT preliminary amendment.			
22.	<input type="checkbox"/>	A substitute specification.			
23.	<input type="checkbox"/>	A change of power of attorney and/or address letter.			
24.	<input type="checkbox"/>	Other items or information:			
"Express Mail Post Office to Addressee" service Mailing Label Number <u>EL541613341US</u>					

U.S. Application No. (If known, use of PCT No.) 097807185	INTERNATIONAL APPLICATION NO. PCT/EP99/07274	ATTORNEY'S DOCKET NUMBER H 3722 PCT/US
17. The following fees are submitted: BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)): Neither International preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO and International Search Report not prepared by the EPO or JPO..... \$1,000.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but International Search Report prepared by the EPO or JPO..... \$860.00 International preliminary examination fee (37 CFR 1.482) not paid to USPTO but international search fee (37 CFR 1.445(a)(2)) paid to USPTO \$710.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) but all claims did not satisfy provisions of PCT Article 33(1)-(4) \$690.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(1)-(4)..... \$100.00		CALCULATIONS PTO USE ONLY
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Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)).		\$ 0
CLAIMS	NUMBER FILED	NUMBER EXTRA
Total Claims	20 - 20 =	0
Independent Claims	3 - 3 =	0
Multiple dependent claims (s)(if applicable)		0
TOTAL OF ABOVE CALCULATIONS		=
Reduction of 1/3 for filing by small entity, if applicable. A Small Entity Statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28).		\$ 0
SUBTOTAL		=
Processing fee of \$130.00 for furnishing the English translation later the <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)).		\$ 0
TOTAL NATIONAL FEE		=
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property		\$ 0
TOTAL FEES ENCLOSED		=
Amount to be: refunded:		\$-----
charged:		\$ 860.00
a. <input type="checkbox"/> A check in the amount of \$_____ to cover the above fees is enclosed. b. <input checked="" type="checkbox"/> Please charge my Deposit Account No. <u>50-1177</u> in the amount of <u>\$860.00</u> to cover the above fees. A triplicate copy of this sheet is enclosed. Order No. <u>01-0208</u> . c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>50-1177</u> . A triplicate copy of this sheet is enclosed. NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137 (a) or (b)) must be filed and granted to restore the application to pending status.		
SEND ALL CORRESPONDENCE TO: Cognis Corporation, Law Dept. 2500 Renaissance Blvd., Suite 200 Gulph Mills, PA 19406		
SIGNATURE: _____ Aaron R. Ettelman NAME ATTORNEY FOR APPLICANT 42.516 REGISTRATION NUMBER		

09/807165

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PATENT

Docket No. H 3722 PCT/US

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

RE: PCT/EP99/07274
International Filing Date: October 1, 1999
Priority Date Claimed: October 10, 1998
Applicant: Daute et al.
Title: ADDITIVES FOR PROCESSING PLASTICS
Applicants' Reference: H 3722 PCT/US

PRELIMINARY AMENDMENT

Commissioner for Patents
Box PCT
Washington, DC 20231

ATTN: DO/EO/US

Prior to the calculation of fees and examination of the above-identified national stage application pursuant to the accompanying submission under 35 U.S.C. §371, please amend the English translation of the International Application submitted herewith, without prejudice, as follows:

In the Specification:

Please amend the instant Specification, without prejudice, as follows:

Please delete all text above line 10, of page 1, and replace the deleted matter with the following new section headings and title of the invention:

--TITLE OF THE INVENTION

**Spheronized Granules, Plastic Additive Compositions
Containing the Same and Methods of Use Therefor**

BACKGROUND OF THE INVENTION--

At page 2, line 5 thereof, please delete the section heading "Description of the

**Preliminary Amendment of U.S. National Stage for International Application
PCT/EP99/07274 filed October 1, 1999**

Invention" and insert the following new section heading and new paragraph:

--BRIEF SUMMARY OF THE INVENTION

The present invention relates generally to additives for the processing of plastics, the additives being in a granular form, characterized in that the granules are present in the form of spherical and/or cylindrical granules with a length-to-diameter ratio of 1:1 to 3:1. The present invention also relates to compositions containing such granules for the processing of plastics and to the use of the granules as auxiliaries for the processing of plastics. --

At page 2, between lines 22 and 23, please add the following new section heading:

--DETAILED DESCRIPTION OF THE INVENTION--

At page 15, between lines 1 and 2, please add the following new paragraph:
--What is claimed is:--

On a separate, new page 16, following page 15, please add the following new section heading and paragraph containing an Abstract of the Disclosure:

--ABSTRACT OF THE DISCLOSURE

Compositions containing at least one plastic additive in granular form wherein the granules are spherical and/or cylindrical with a length-to-diameter ratio of from 1:1 to 3:1; are disclosed. Processes for preparing such granular compositions, and methods of stabilizing plastics during processing using such compositions are also disclosed.--

In the Claims:

Please add new claims 6-25, as follows:

--6. (New) A composition comprising granules containing at least one plastic additive, the granules having a length-to-diameter ratio of 1:1 to 3:1.--

**Preliminary Amendment of U.S. National Stage for International Application
PCT/EP99/07274 filed October 1, 1999**

--7. (New) The composition according to claim 6, wherein the granules have a diameter of from 0.5 to 5 mm.--

--8. (New) The composition according to claim 6, wherein the granules have a diameter of from 0.8 to 3 mm.--

--9. (New) The composition according to claim 6, wherein the granules have a uniform size and shape.--

--10. (New) The composition according to claim 6, wherein the granules are substantially spherical.--

--11. (New) The composition according to claim 6, wherein the granules have a length-to-diameter ratio of 1:1.--

--12. (New) The composition according to claim 6, wherein the at least one plastic additive comprises a component selected from the group consisting of lubricants, stabilizers, and mixtures thereof.--

--13. (New) The composition according to claim 12, wherein the component is selected from the group consisting of calcium soaps, zinc soaps, and mixtures thereof.--

--14. (New) The composition according to claim 6, wherein the granules are substantially spherical and have a diameter of from 0.8 to 3 mm, and wherein the at least one plastic additive comprises a component selected from the group consisting of calcium soaps, zinc soaps, and mixtures thereof.--

--15. (New) A granular composition prepared by a process comprising:

(a) providing cylindrical granules of a composition containing at least

**Preliminary Amendment of U.S. National Stage for International Application
PCT/EP99/07274 filed October 1, 1999**

one plastic additive; and

(b) spheronizing the cylindrical granules to form granules having a length-to-diameter ratio of from 1:1 to 3:1.--

--16. (New) The granular composition according to claim 15, wherein providing the cylindrical granules comprises: (i) extruding a composition containing at least one plastic additive into a fine strand; and (ii) cutting the fine strand into cylindrical granules.--

--17. (New) The granular composition according to claim 16, wherein the composition is extruded using a twin-screw extruder.--

--18. (New) The granular composition according to claim 17, wherein the composition is extruded at a temperature of from 20 to 110°C and a pressure of from 25 to 60 bar.--

--19. (New) The granular composition according to claim 15, wherein spheronizing is accomplished using a spheronizer having a rotating bottom disk.--

--20. (New) The granular composition according to claim 19, wherein spheronizing is performed at a rotational speed of 320 rpm with a residence time of 30 seconds.--

--21. (New) The granular composition according to claim 15, wherein the process further comprises impregnating the granules with an additional active substance.--

--22. (New) The granular composition according to claim 21, wherein impregnating the granules with an additional active substance is accomplished via surface-powdering.--

**Preliminary Amendment of U.S. National Stage for International Application
PCT/EP99/07274 filed October 1, 1999**

--23. (New) The granular composition according to claim 22, wherein the surface powdering is carried out at least partly during spheronizing.--

--24. (New) The granular composition according to claim 18, wherein spheronizing is carried out using a spheronizer having a rotating bottom disk operating at a rotational speed of 320 rpm with a residence time of 30 seconds; and the process further comprises impregnating the granules with an additional active substance.--

--25. (New) A method of stabilizing a plastic composition during processing, said method comprising:

- (a) providing a plastic composition;
- (b) providing a granular composition comprising granules containing at least one plastic additive, the granules having a length-to-diameter ratio of from 1:1 to 3:1; and
- (c) combining the plastic composition and the granular composition prior to processing completion.--

Please cancel claims 1-5, without prejudice.

REMARKS

Claims 6-25 are currently pending in the instant application.

The Specification has been amended to delete the original section headings and to insert the preferred section headings pursuant to 37 C.F.R. §1.77. A new Title of the Invention has been inserted. An Abstract of the Disclosure in accordance with the abstract of the corresponding international publication has been added on a separate sheet following the claims. It is submitted that the amendments to the Specification made herein introduce no new matter. All of the amendments to the Specification constitute deletions of original section headings and/or paragraphs, and insertions or additions of new section headings and/or paragraphs. Accordingly, pursuant to 37 C.F.R. §1.121(b)(1)(iii), no separate page captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE" is necessary. A separate page containing a clean copy of the Abstract of the Disclosure has been added for the Examiner's convenience. Entry of the amendments to the Specification made herein are therefore proper and respectfully requested.

Original claims 1-5 have been canceled and replaced with new claims 6-25 solely for the purpose of improving clarity and grammar, which may suffer in translation, and not for any reason which relates to the statutory requirements for a patent. New claims 6-25 have not been added in response to any rejection, nor in anticipation of any rejection. Applicants respectfully submit that the scope of new claims 6-25 generally corresponds to the scope of original claims 1-5, and that new claims 6-25 are no narrower than original claims 1-5. Furthermore, although a moot point in view of their cancellation, Applicants respectfully submit that original claims 1-5 satisfied the requirements of 35 U.S.C. §112, as filed. New claims 6-25 are supported by the claims as originally filed and in the Specification, for example, at page 2, lines 19-22; at page 7, lines 4-16; at page 8, lines 3-23; and in the Examples. No new matter has been introduced. All of the amendments to the Claims constitute cancellation of original claims and the addition of new claims. Accordingly, pursuant to 37 C.F.R. §1.121(c)(1)(ii), no separate page captioned "VERSION WITH MARKINGS TO SHOW CHANGES MADE" is necessary. Entry is therefore proper and respectfully requested.

**Preliminary Amendment of U.S. National Stage for International Application
PCT/EP99/07274 filed October 1, 1999**

Prompt examination of the instant application in view of the amendments made
herein is respectfully requested.

Respectfully submitted,

PETER DAUTE, et al.

April 14, 2001
(Date)

A. R. Ettelman
AARON R. ETTELMAN
(Reg. No. 42,516)
Attorney for Applicants
Telephone: (610) 278-4930
Facsimile: (610) 278-6548
E-Mail: AARON.ETTELMAN@HENKEL-AMERICAS.COM

Cognis Corporation, Patent Dept.
2500 Renaissance Blvd., Suite 200
Gulph Mills, PA 19406

ARE/ras

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ABSTRACT OF THE DISCLOSURE

Compositions containing at least one plastic additive in granular form wherein the granules are spherical and/or cylindrical with a length-to-diameter ratio of from 1:1 to 3:1; are disclosed. Processes for preparing such granular compositions, and methods of stabilizing plastics during processing using such compositions are also disclosed.

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Additives for Processing Plastics

Field of the Invention

This invention relates to additives for the processing of plastics, the additives being present in a granular supply form, characterized in that the granules are present in the form of spherical to cylindrical granules with a
5 length-to-diameter ratio of 1:1 to 3:1. The invention also relates to compositions containing such granules for the processing of plastics and to the use of the granules as auxiliaries for the processing of plastics.

Prior Art

10 Plastics are polymers which consist structurally of molecule chains with numerous, almost infinitely recurring structural elements and which differ in their physical properties from low molecular weight compounds. Thus, polymers have high tensile strengths and elasticity, for example, by comparison with low molecular weight compounds.

15 In the course of industrial processing, suitable additives are incorporated in polymerized plastic melts to facilitate their further processing and their intended application. In this way, basic polymers can be adapted to meet individual requirements in regard to the required properties.

20 Additives for plastics are available in various supply forms. For solid compounds, the simplest supply form is the powder. Unfortunately, **powder** can cake too easily and leads to dust emissions during processing. Compression of the powder in a granulator gives **rodlet granules**. Although the dust component is small, processing problems can be caused
25 by the poorer handling behavior of such granules and the dust present. For some additives which have a high meltable component, **pellets** or **flakes** are available as supply forms. For production, the particular product

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is melted and then cooled down on cooled flaking rollers or belts. Pellets are dust-free and may readily be incorporated in the plastic. However, thermally labile or reactive compounds cannot be flaked or pelleted.

5

Description of the Invention

The problem addressed by the present invention was to provide additives for the processing of plastics which would be easy to handle and which could readily be incorporated in thermoplastics.

10 It has now been found that extruded plastic additives can be converted by spheronizing, such as "marumerizing", into a substantially spherical or cylindrical shape. Granules thus shaped ("**beads**") on the one hand afford the advantage of easy handling and freedom from dust; on the other hand, they can readily be incorporated during the processing of thermoplastics, i.e. dispersed in the plastics.

15 The additive beads thus produced are eminently suitable as a supply form for plastic additives, more particularly for stabilizer and/or lubricant compounds for the processing of PVC, for example those based on calcium/zinc and on lead.

20 The present invention relates to additives for the processing of plastics, the **additives being present in a granular supply form**, characterized in that the granules are present in the form of spherical or cylindrical granules with a length-to-diameter ratio of 1:1 to 3:1.

25 With regard to the length-to-diameter ratio, attention is drawn to the following: the additives granules according to the invention assume the form of bodies of rotation (i.e. they may also be graphically referred to as "beads"). The length of the longitudinal axis of the beads is termed the "length" while the maximum diameter of the beads (as measured perpendicularly of the longitudinal axis) is termed the "diameter". In cases where the length-to-diameter ratio is 1:1, the beads are spheres. In the
30 process described hereinafter for producing the additive granules, the bead

09807165-070201

form is obtained by virtue of the fact that, after extrusion and chopping up of the extruded strands at the granulation head with multiple-bore dies, cylindrical particles are initially obtained and are then rounded off at their ends in spheronizing machines.

- 5 The supply form according to the invention for the plastic additives combines several favorable applicational properties which are not found in this number in conventional supply forms, such as powders or pellets. For experimental proof, reference is made to the Examples. As mentioned above, these positive applicational properties include excellent transport
- 10 behavior and the absence of dust - both properties which guarantee excellent handling behavior. In addition, the supply form according to the invention opens up extensive possibilities in regard to the "range of formulation". By this is meant that the beads according to the invention may contain not just one additive, but also several additives irrespective of
- 15 whether these additives are - for instance - thermally labile, infusible or substantially infusible, represent otherwise difficult-to-handle polymers, etc. The positive applicational properties mentioned remain intact even when the formulation range of the beads according to the invention is broad, i.e. when several different additives, including thermally labile and otherwise
- 20 difficult-to-handle additives, are present in the beads.

According to the invention, the **plastics** may be selected basically as required from the thermoplastics known to the relevant expert. The following are examples of suitable plastics:

- homopolymers of an α -olefin containing two to eight carbon atoms,
- 25 copolymers of two corresponding α -olefins, preferably copolymers of ethylene, ethylene homopolymers, such as HDPE (high-density polyethylene), LDPE (low-density polyethylene), VLDPE (very low-density polyethylene), LLDPE (linear low-density polyethylene), MDPE (medium-density polyethylene), UHMPE (ultra-high-molecular
- 30 polyethylene), VPE (crosslinked polyethylene), HPPE (high-pressure

polyethylene), isotactic polypropylene, syndiotactic polypropylene, Metallocen-catalyzed polypropylene, high-impact polypropylene, random copolymers based on ethylene and propylene, block copolymers based on ethylene and propylene, homopolymers based on 1-butylene, 1-pentylene, 1-hexylene, 1-octylene, isobutylene, 2-methyl-1-butylene, 3-methyl-1-pentylene, 4-methyl-1-pentylene, 2,3-dimethyl-1-butylene, 2-ethyl-1-butylene and mixtures thereof.

- copolymers of ethylene with 1-butylene, 1-hexylene, 1-octylene and 4-methyl-1-pentylene.
- ethylene/vinyl acetate copolymers, ethylene/ethyl acetate copolymers, ethylene/acrylic acid copolymers and mixtures thereof.
- ethylene/propylene rubber (EPDM), including diene-modified types (EPR), styrene/butadiene/styrene copolymers (SBS), styrene/ethylene/butylene/styrene copolymers (SEBS) and mixtures thereof.
- halogen-containing plastics, more particularly homopolymers of vinyl compounds, for example vinyl chloride.

According to the invention, the **additives** may be selected basically as required from the additives known to the relevant expert for the processing of thermoplastics. Examples of suitable additives are antistatic agents, antifogging agents, antioxidants, UV stabilizers, coupling agents, calendering aids, mold release agents, lubricants, release agents, slip agents, plasticizers, perfumes, flame retardants, fillers and agents for increasing thermal stability (heat stabilizers).

So far as the terms "lubricants" and "release agents" are concerned, attention is drawn to the following: in the standard language of the expert, release agents are products which reduce the frictional resistances primarily between the polymer melt and the steel surface of the machine used for molding; the effect of reducing the frictional resistance is that the

melt pressure of the melt is reduced. By contrast, lubricants act predominantly in the polymer melt and reduce the internal frictional forces so that, even with high filler contents, the melt retains good plastic flow which is important for filling the mold.

- 5 In one embodiment of the invention, calcium salts and/or magnesium salts and/or aluminium salts and/or zinc salts solid or liquid at 20°C selected from
- a) calcium salts of saturated or unsaturated, linear or branched monocarboxylic acids containing 6 to 36 carbon atoms,
 - 10 b) calcium salts of unsubstituted or C₁₋₄-alkyl-substituted benzoic acid,
 - c) zinc salts of saturated or unsaturated, linear or branched monocarboxylic acids containing 6 to 36 carbon atoms,
 - d) magnesium salts of saturated or unsaturated, linear or branched monocarboxylic acids containing 6 to 36 carbon atoms,
 - 15 e) magnesium salts of saturated or unsaturated dicarboxylic acids containing 6 to 10 carbon atoms,
 - f) aluminium salts of saturated or unsaturated, linear or branched monocarboxylic acids containing 6 to 36 carbon atoms
- are used as **lubricants or release agents**.

- 20 The above-mentioned calcium, magnesium, zinc and aluminium salts may be used both individually and in admixture with one another.

- Other lubricants or release agents which may be used individually or in combination with one another are the relevant substances known from the prior art. Compounds of the following types are preferably used:
- 25 hydrocarbon waxes which melt at temperatures of 70 to 130°C, oxidized polyethylene waxes, free fatty acids containing 8 to 22 carbon atoms and branched-chain isomers thereof, for example stearic acid or even hydroxystearic acid, α -olefins, wax esters, i.e. esters of relatively long-chain monocarboxylic acids and monoalcohols, primary and secondary,
 - 30 saturated and unsaturated higher alcohols preferably containing 16 to 44

carbon atoms in the molecule, ethylenediamine stearate, montanic acid esters of diols, for example of ethanediol, butane-1,3-diol and glycerol, mixtures of such montanic acid esters with nonesterified montanic acids, partial esters of fatty acids containing 8 to 22 carbon atoms and polyols containing 2 to 6 carbon atoms and 2 to 6 hydroxyl groups which contain on average at least one free polyol hydroxyl group per molecule. Also suitable are the mixed esters described in **DE-C-19 07 768** with hydroxyl or acid values of 0 to 6 of aliphatic, cycloaliphatic or aromatic dicarboxylic acids containing 2 to 22 carbon atoms in the molecule, aliphatic polyols containing 2 to 6 hydroxyl groups in the molecule and aliphatic monocarboxylic acids containing 12 to 30 carbon atoms in the molecule. Examples of these mixed esters are mixed esters of maleic acid/pentaerythritol/behenic acid, mixed esters of adipic acid/pentaerythritol/oleic acid and mixed esters of adipic acid/pentaerythritol/stearic acid. According to the invention, corresponding lubricants or release agents may be used both individually and in combination with one another and also in combination with the above-mentioned calcium, magnesium or aluminium salts.

In one embodiment of the invention, compounds selected from the group consisting of talcum, kaolin, chalk and the like, are used as **fillers**.

In another embodiment of the invention, compounds selected from the group consisting of calcium and/or zinc soaps, antioxidants, cationic layer compounds, for example hydrotalcites or modified hydrotalcites, zeolites, are used as **heat stabilizers**.

As known to the expert, additives used in the processing of plastics may be classified not only in regard to their function but also in regard to their chemical structure. From the structure perspective, too, the additives according to the invention are not subject to any restrictions. It is pointed out in this connection that compounds belonging to a certain class, i.e. compounds which may be structurally assigned to the same class, often perform not just one but two or more functions in practice. For example,

calcium or zinc soaps - as described above - may act as lubricants and/or release agents but may also be used to improve thermal stability, for example in the processing of polyvinyl chloride (PVC).

The additive granules ("beads") according to the invention are generally **produced** as follows: a composition containing plastic additives, more particularly a composition consisting solely of plastic additives, is introduced into a single-screw or twin-screw extruder. Twin-screw kneader/extruders (twin-screw co-rotating or contra-rotating extruders) are preferred, an extrusion pressure of 15 to 100 bar and more particularly in the range from 25 to 60 bar preferably being established. Basically, the temperature in the extruder is not critical although extrusion is preferably carried out at 20 to 110°C. The extruders used are provided with granulation heads equipped in particular with multiple-bore extrusion dies. The effect of this is that the melt is extruded into fine strands which, after leaving the die, are converted into cylindrical granules by means of a chopping blade. Multiple bore dies with bores between 0.8 and 5 mm and more particularly between 1.5 and 2.5 mm in diameter are preferably used. The chopping temperature is preferably adjusted to a value of 30 to 110°C and more preferably to a value of 40 to 80°C. These granules are then spheronized continuously or in batches in conventional spheronizers. The spheronizing temperature is adjusted so that the granules have sufficient plasticity. The preferred spheronizing temperature is between 20 and 110°C. Relevant spheronizing techniques, especially marumerizing, are known to the expert. Reference is made by way of example in this connection to the disclosure of German patent **DE-C-12 94 351** "Vorrichtung zur Herstellung von kugelförmigen Körnern aus feuchten, vorgeformten Teilchen (arrangement for the production of spherical granules from moist preformed particles)" which describes a spheronizer with a rotating bottom disk, the required degree of spheronizing being adjustable by variation of the residence time of the granules in the

spheronizer and/or the rotational speed of the disk. According to the invention, any commercially available spheronizers may be used.

In one embodiment, the still plastic additive granules initially formed may be impregnated with other active substances before, during or after spheronizing. These other active substances may be, for example, particularly heat-sensitive plastic additives. In one particularly preferred embodiment, the additive granules according to the invention are surface-**powdered** with one or more compounds. Organic active substances, inorganic active substances and release agents, which may be used individually or in combination with one another, are particularly suitable for this purpose. Zeolites and cationic layer lattice compounds, for example hydrotalcites or modified hydrotalcites, are most particularly suitable. The compounds used for powdering are used in particular in quantities of 0.01 to 5% by weight, based on the additive granules (beads). Powdering is preferably carried out at the same time as spheronizing.

As already mentioned, the plastic additive granules ("beads") according to the invention are present in the form of spherical or cylindrical granules with a length-to-diameter ratio of 1:1 to 3:1. In one important embodiment, particle size and particle shape are uniform. The preferred particle form is spherical. Preferred particle sizes are in the range from 0.5 to 5 mm sphere diameter and more particularly in the range from 0.8 to 3 mm. The constituents of these spheres may correspond in type and quantity to standard formulations for plastic additive compositions.

In another embodiment, the additive granules according to the invention may be recycled. By this is meant that they may be reused together with other substances in the first process step for producing the additive granules - extrusion.

The present invention also relates to **compositions** for the processing of plastics containing additives for the processing of plastics, characterized in that the compositions are present in a granular supply form

and in that the granules are present in the form of spherical or cylindrical granules with a length-to-diameter ratio of 1:1 to 3:1.

The present invention also relates to the **use** of granules as auxiliaries for the processing of plastics, characterized in that the granules consist of one or more typical plastic additives and are present in the form of spherical or cylindrical granules with a length-to-diameter ratio of 1:1 to 3:1.

The present invention also relates to a **process** for the production of plastics, characterized in that powder-form and/or granular thermoplastic homopolymers and/or copolymers are subjected to molding, more particularly extrusion, injection molding or film blowing, together with the additive granules according to the invention. Any of the machines known to the relevant expert may be used, the various commercially available single- and twin-screw extruders being particularly suitable for extrusion.

Examples

1. Materials

For the following tests, a commercially available powder-form Ca/Zn stabilizer ("Stabilox CZ 2697", Henkel, Düsseldorf) was processed

- to rodlet granules (= additive granules intended for comparison) and
- to spheronized beads (= additives granules according to the invention).

2. Production of the materials

2.1. Rodlet granules

A CPM granulator (pressing temperature 30°C) was used to produce the rodlet granules. The rodlets obtained had a diameter of 3 mm and a length of about 5 mm.

2.2. Beads

The additive granules according to the invention were produced as

follows: powder-form Stabinox CZ 2697 was continuously added to a twin-screw kneader (Werner & Pfleiderer type ZSK 35 extruder = twin-screw kneader/co-rotating extruder; screw diameter = 25 mm; L/D = 12) of which the housing, including the granulation head of the extruder, was heated to 50°C. The screw speed was 160 r.p.m., the extrusion pressure 42 bar and the specific energy input 0.017 kWh/kp. The melt was extruded through the extruder head multiple-bore die to form fine strands which, after leaving the die, were size-reduced to cylindrical granules by a chopping blade (hot-chopping granulation head with a 47-bore die (bore diameter 2 mm); twin-bladed cutter; chopping temperature 50°C; blade speed 2650 r.p.m.). The granules collecting from the extruder/granulator were spheronized in a commercially available spheronizer of the Marumerizer type (Schlüter RM 400 Spheronizer - spheronizing conditions: rotational speed 320 r.p.m.; residence time 30 s). In the spheronizer, the granules were powdered with 3% by weight - based on the Stabinox CZ 2697 used - of Sasil A 40 (Wessalith P, Degussa AG). The granules were then sieved through a 2.5 mm sieve, about 10% of oversize granules (agglomerates) being removed. The additive granules obtained ("beads") were substantially spherical.

3. Performance tests

3.1. Determination of abrasion

Quantities of 50 g of the rodlet-like and the bead-like Ca/Zn stabilizer (produced as described above in 2.1. and 2.2.) were subjected to mechanical stressing in a Retsch sieve analyzer and the abrasion was determined. The following parameters were adjusted: time 5 mins., vibration amplitude 1 mm, mesh width of test sieve: 0.315 mm.

Abrasion

<i>Supply Form</i>	<i>Abrasion [g]</i>	<i>Abrasion [g]</i>
Rodlet granules 2.1.	0.23	0.46
Beads 2.2.	0.02	0.04

As can be seen from the Table, the beads showed considerably lower abrasion. This is of great importance from the applicational perspective.

3.2. Determination of flowability

To determine flowability, quantities of 96 g of the rodlet granules 2.1. and the beads 2.2. were introduced into a funnel closed at its lower end, the outlet was opened and the outflow time was measured. The funnel (polyethylene powder funnel with an upper diameter of 105 mm) had the following dimensions: diameter of outlet: 23 mm, length of outlet: 23 mm).

Flowability

<i>Supply form</i>	<i>Outflow time [s]</i>
Rodlet granules	2.6 / 3.2 / 2.8
Beads	1.5 / 1.4 / 1.4

The outflow times were measured three times.

As can be seen from the Table, the outflow time was significantly shorter for the beads according to the invention than for the rodlet granules. In practice, this means that flowability is considerably improved, thus ensuring higher feed rates.

3.3 Use in PVC formulations

The performance properties of powder, rodlet granules and beads

were compared using the Ca/Zn stabilizer Stabilox CZ 2697. To this end, the individual supply forms were processed with polyvinyl chloride (PVC, "Evipol SH 6830" = commercially available suspension PVC) in a Herschel mixer to form a dry blend (quantity of material: 3 kg, heating to 120°C, subsequent cooling)

Formulations

	<i>B1</i>	<i>B2</i>	<i>B3</i>
EVIPOL SH 6830	100	100	100
Stabilox CZ 2697 (powder 1.)	3.35	-	-
Stabilox CZ 2697 (rodlet granules 2.1.)	-	3.35	-
Stabilox CZ 2697 (beads 2.2.)	-	-	3.35

Figures in Table = parts by weight

Formulations B1 to B3 (dry blends) were tested first for sieve residue and bulk density:

Sieve residue and bulk density

	<i>Sieve residue [>0.5 mm]</i>	<i>Bulk density</i>
Formulation B1	0.09	621
Formulation B2	0.12	617
Formulation B3	0.07	614

Sieve residue: expressed in g based on 200 g of dry blend

Bulk density: expressed in g/l

Explanation of the determination of the sieve residue: as an indicator of the degree of dispersion during mixing, 200 g of the dry blend

were sieved through a 0.5 mm mesh sieve. All the particles above 0.5 mm in size remain in the sieve in this test. This is known as the sieve residue.

The dry blends were then extruded to a flat strip in a Weber twin-screw extruder. The extrusion parameters were as follows: screw speed 20 r.p.m., machine load 42, temperature 180°C. The strip had a thickness of 1.5 mm and a width of 25 mm.

The color of the strips was determined immediately after their production (so-called initial color) using the L*,a*,b* method known to the expert (cf. DIN 6174, CIELAB 1976). The b* value indicates the position on the blue/yellow axis. Normally, the b* value is also called the yellow value. A commercially available instrument known as a "Micro Color" (Dr. Lange) was used for the measurements. The color values are set out in the following Table. Another parameter determined was how long it took the test strips to turn black in color during heat treatment in a "thermo-oven" (cf. DIN 5033). To this end, the strips were heated at 180°C in a thermo-oven, being briefly removed from the oven every 15 minutes for visual examination. The time in minutes which the strips took to turn black is termed "long-term stability". The test results are set out in the following Table.

Heat stability

	Yellow value b* after		Long term stability (= end of stability after)
	0 mins.	15 mins.	
Formulation B1	15	34	90 mins
Formulation B2	15	35	90 mins.
Formulation B3	15	35	90 mins.

Overall, it was found that the strip which had been produced using

the beads according to the invention (formulation B3) was as heat-stable as the strips which had been produced using powder or rodlet granules (formulations B1 and B2). Accordingly, the above-mentioned performance-related advantages of the additive granules according to the invention ("beads"), i.e. in particular less dust, better flowability, greater formulation flexibility, have no adverse effects on their ability to heat-stabilize PVC.

030745-07004

CLAIMS

1. Additives for the processing of plastics, the additives being present in a granular supply form, characterized in that the granules are present in the form of spherical or cylindrical granules with a length-to-diameter ratio of 1:1 to 3:1.
- 5 2. Additives as claimed in claim 1, characterized in that the surface of the granules is additionally powdered with inorganic and/or organic active substances and/or release agents.
3. Compositions for the processing of plastics containing granules according to claim 1 or 2.
- 10 4. The use of granules as auxiliaries for processing plastics, characterized in that the granules consist of one or more typical plastic additives and are present in the form of spherical or cylindrical granules with a length-to-diameter ratio of 1:1 to 3:1.
- 15 5. A process for the production of plastics, characterized in that powder-form and/or granular thermoplastic homopolymers and/or copolymers are subjected to molding together with the additive granules according to claim 1 or 2.

000745-070204

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Rev. 9/95

U.S. Department of Commerce
Patent and Trademark Office

DECLARATION FOR UTILITY OR DESIGN PATENT APPLICATION

☐ Declaration Submitted with Initial Filing OR ☒ Declaration Submitted after Initial Filing

Attorney Docket Number

H 3722 PCT/US

First Named Inventor

DAUTE, Peter

COMPLETE IF KNOWN

Application Number

09/807,165

Filing Date

07/02/2001

Group Art Unit

Examiner Name

As a below named inventor, I hereby declare that:

My residence, post office address, and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

ADDITIVES FOR PROCESSING PLASTICS

the specification of which

(Title of the Invention)

☐ is attached hereto

OR

☒ was filed on (MM/DD/YYYY)

10/01/1999

as United States Application Number or PCT International

Application Number

PCT/EP99/07274

and was amended on (MM/DD/YYYY)

(if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment specifically referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in Title 37 Code of Federal Regulations, § 1.56.

I hereby claim foreign priority benefits under Title 35, United States Code §110(a)-(d) or §365(b) of any foreign application(s) for patent or inventor's certificate, or §365(e) of any PCT International application which designated at least one country other than the United States of America, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or of any PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application Number(s)	Country	Foreign Filing Date (MM/DD/YYYY)	Priority Not Claimed	Certified Copy Attached? YES NO
198 46 774.5	DE	10/10/1998	<input type="checkbox"/>	<input checked="" type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>
			<input type="checkbox"/>	<input type="checkbox"/>
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			<input type="checkbox"/>	<input type="checkbox"/>
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☐ Additional foreign application numbers are listed on a supplemental priority sheet attached hereto:

I hereby claim the benefit under Title 35, United States Code §119(e) of any United States provisional application(s) listed below.

Application Number(s)	Filing Date (MM/DD/YYYY)	Additional provisional application numbers are listed on a supplemental priority sheet attached hereto.
		<input type="checkbox"/>

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DECLARATION

Page 2

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s), or §365(c) of any PCT international application, designating the United States of America, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT international application in the manner provided by the first paragraph of Title 35, United States Code §112.1 acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations §1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application.

U.S. Parent Application Number	PCT Parent Number	Parent Filing Date (MM/DD/YYYY)	Parent Patent Number (if applicable)
	PCT/EP99/07274	10/01/1999	

☐ Additional U.S. or PCT international application numbers are listed on a supplemental priority sheet attached hereto.

As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

☐ Firm Name Customer Number or label

☒ List Attorney(s) and/or agent(s) name and registration number below:

Name	Registration Number	Name	Registration Number
John E. Drach Steven J. Trzaska	32,891 36,296	Aaron R. Ettelman Henry E. Millson, Jr.	42,516 18,980

☐ Additional attorney(s) and/or agent(s) named on a supplemental sheet attached hereto.

Please direct all correspondence to: ☒ Customer Number or label **23657** OR ☒ Fill in correspondence address below

Name **Aaron R. Ettelman**

Address

Address

City State ZIP

Country Telephone **610-278-4930** Fax **610-278-6548**

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Name of Sole or First Inventor: ☐ A petition has been filed for this unassigned

Given Name **Peter** Middle Initial Family Name **Daute** Suffix e.g. Jr.

Inventor's Signature **Peter Daute** Date **2004-05-22**

Residence: **Beverstedt** **CEX** State Country **Germany** Citizenship **Germany**

Post Office Address **Adolf-Butenandt-Strasse 25a**

Post Office Address

City **27616 Beverstedt** State Zip Country **Germany** Applicant Authority

☒ Additional inventors are being named on supplemental sheet(s) attached hereto

DECLARATION

ADDITIONAL INVENTOR(S)
Supplemental Sheet

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name	Wolfgang	Middle Initial	Family Name	Seiter	Suffix e.g. Jr.
Inventor's Signature	<i>Wolfgang Seiter</i>				Date
Residence: City	Neuss	State	Country	Germany	Citizenship
Post Office Address	Lorbeerweg 7				
Post Office Address					
City	41469 Neuss	State	Zip	Country	Germany
				Applicant Authority	

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name	Joerg-Dieter	Middle Initial	Family Name	Klamann	Suffix e.g. Jr.
Inventor's Signature	<i>Joerg-Dieter Klamann</i>				Date
Residence: City	Bremerhaven	State	Country	Germany	Citizenship
Post Office Address	Im Schiffsmoor 8d				
Post Office Address					
City	27674 Bremerhaven	State	Zip	Country	Germany
				Applicant Authority	

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name	Peter	Middle Initial	Family Name	Wedl	Suffix e.g. Jr.
Inventor's Signature	<i>Peter Wedl</i>				Date
Residence: City	Bremerhaven	State	Country	Germany	Citizenship
Post Office Address	Loeningstrasse 13				
Post Office Address					
City	27668 Bremerhaven	State	Zip	Country	Germany
				Applicant Authority	

Name of Additional Joint Inventor, if any:

☐ A petition has been filed for this unsigned inventor

Given Name		Middle Initial	Family Name		Suffix e.g. Jr.
Inventor's Signature					Date
Residence: City		State	Country		Citizenship
Post Office Address					
Post Office Address					
City		State	Zip	Country	Applicant Authority

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